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Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL)

Part 1. Description of Tritium Dose Model (DCART) for Chronic Releases from LLNL

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DISCLAIMER

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ABSTRACT

DCART (Doses from Chronic Atmospheric Releases of Tritium) is a spreadsheet model developed at Lawrence Livermore National Laboratory (LLNL) that calculates doses from inhalation of tritiated hydrogen gas (HT), inhalation and skin absorption of tritiated water (HTO), and ingestion of HTO and organically bound tritium (OBT) to adult, child (age 10), and infant (age 6 months to 1 year) from routine atmospheric releases of HT and HTO. DCART is a deterministic model that, when coupled to the risk assessment software Crystal Ball®, predicts doses with a 95th percentile confidence interval. The equations used by DCART are described and all distributions on parameter values are presented. DCART has been tested against the results of other models and several sets of observations in the Tritium Working Group of the International Atomic Energy Agency's Biosphere Modelling and Assessment Programme. The version of DCART described here has been modified to include parameter values and distributions specific to conditions at LLNL. In future work, DCART will be used to reconstruct dose to the hypothetical maximally exposed individual from annual routine releases of HTO and HT from all LLNL facilities and from the Sandia National Laboratory's Tritium Research Laboratory over the last fifty years.

INTRODUCTION

Historically, more tritium has been released from Lawrence Livermore National Laboratory (LLNL) over its fifty-year history than any other radionuclide; as much as $3.5\ 10^{16}$ Bq (close to one million Ci), primarily in the form of tritiated gas (HT), may have been released. The radiological dose consequences of these releases are thought to be of no health concern (ATSDR 2002). Nevertheless, a careful analysis of annual doses that accounts for uncertainties in source terms and all pathways to dose should be carried out to determine what doses are most likely to have been received by a member of the public.

A model has been developed at LLNL to estimate tritium doses from routine operations at LLNL and neighboring Sandia National Laboratory. This model, DCART (**D**oses from **C**hronic **A**tmospheric **R**eleases of **T**ritium), is a deterministic spreadsheet code in Microsoft Excel®. When coupled to Crystal Ball 2000®[1], a risk analysis software package that provides uncertainty and sensitivity analyses for spreadsheet codes, DCART produces probabilistic results. The use of DCART to predict historical tritium doses will provide a set of consistent, defensible dose estimates based on the latest knowledge of tritium transfer through the environment. Given the history of tritium dose predictions at LLNL (below), a new model and new approach is clearly needed.

Doses to the public from tritium and other radionuclides released to the environment from operations at the Lawrence Livermore National Laboratory (LLNL) are reported annually in the site annual environmental reports (e.g., Sanchez et al. 2003). Site environmental reports date back to 1959, when they were addressed to the United States Atomic Energy Commission; site environmental reports have been made available to the public since 1971. In 1974, the first year doses calculated from source terms were published in an annual report, the meteorological dispersion model used for the calculations was based on the model of Pasquill modified by Gifford and Hilsmeir (1962). From 1975 through 1991, as required for reporting purposes by the Department of Energy (DOE), doses were calculated using the Continuous-Point-Source (CPS) Code (Peterson et al. 1976). From 1986 to the present, as mandated by the Environmental Protection Agency (EPA) for compliance with National Emission Standards for Hazardous Air Pollutants (NESHAPs), 40 CFR 61 Subpart H (Office of the Federal Register 1986), the AIRDOS-EPA Clean Air Act Code (CAAC) (Moore et al. 1979; USEPA 1989) was used to predict dose to the public. Since 1992, the dispersion model used to calculate doses from releases of tritium to the atmosphere at LLNL has been the direct descendent to AIRDOS-EPA, CAP88-PC (Parks 1992).

From 1986 to 1991, doses reported in the LLNL annual environmental reports were calculated using both AIRDOS-EPA and the CPS Code for continuity with previous years. The major difference between the two codes is that only inhalation is calculated by the CPS code, while both inhalation and ingestion doses are calculated by AIRDOS-EPA. Thus doses from the CPS code are

[1] Decisioneering, Inc. 1515 Arapahoe Street, Suite 1311, Denver Colorado USA 80202.

expected to be lower, presumably by a reasonably consistent fraction, than those of AIRDOS-EPA (Table 1). However, in 1986 and 1987, the tritium doses from the CPS code were actually higher than those of AIRDOS-EPA. In 1986, LLNL was "unable to use detailed exposure data for the LLNL sites. A variety of assumptions and approximations were necessary in order to complete the calculations. The doses are considered estimates" (Holland et al. 1987). In 1987, LLNL had "not yet fully tailored the CAAC (i.e., AIRDOS-EPA) to site-specific exposure and meteorological parameters. For this reason, the CAAC was run by Oak Ridge National Laboratory using a variety of assumptions, which were necessary to complete the calculations" (Holland and Brekke 1988). After 1988, presumably the same source terms were used for each model and the location of the maximally exposed individual would have been the same, yet inconsistent differences are observed in the doses. These differences lead to questions, such as "Are the differences entirely due to dispersion modeling?" and "Were some assumptions used one year for one model not used another year for the same model?". Short of duplicating the calculations, there is no way to determine what caused the differences between the two sets of predictions. These differences highlight the importance of using DCART with consistent and well-documented assumptions to ensure that calculated doses are truly comparable over time.

Table 1. Maximum offsite dose in mrem from HTO released from the Tritium Facility at LLNL. Numbers were obtained from LLNL annual environmental reports (UCRL-50027-yr).

	Clean Air Act Code (AIRDOS-EPA)	Continuous Point Source (CPS) Code
1986	0.03	0.04
1987	0.07	0.12
1988	0.55	0.10
1989	0.52	0.26
1990	0.22	0.099
1991	0.08	0.065

Until 1998, all reported LLNL doses from routine atmospheric tritium releases were based solely on monitored quantities of tritiated water (HTO) released to the atmosphere; releases of tritiated hydrogen gas (HT) were ignored. This approach was based on the belief that HT is biologically inert and 25,000 times less radiotoxic than HTO (ICRP 1979). For 1998, the EPA (Region IX) mandated that LLNL's tritium doses be modeled as if all released tritium were HTO. This decision was based on the fact that the model mandated for regulatory compliance (CAP88-PC, Parks 1992, 1997) calculates dose from HTO only and the recognition that HT converts quite rapidly to HTO in soil when the HT plume comes to ground (McFarlane et al. 1978; Brown et al. 1988). Experimental data under chronic conditions (Davis and Bickel 2000) have shown that no more than about 10% of the HT at any location gets converted to HTO. The dose consequences of this conversion are a bit higher than 10%, as will be discussed later. Nevertheless, treating all released HT as if it were HTO may grossly overestimate dose.

Only recently have doses predicted at LLNL accounted for ingesting organically bound tritium (OBT) in foods, and no regulatory model as of 2003 includes dose from OBT. The dose from ingesting 1 Bq of OBT is about 2.3 times that from ingesting 1 Bq of HTO (ICRP 1996) because OBT has a longer biological half-life than does HTO. Therefore, the contribution of OBT to dose should be included in dose calculations.

At present, a few published steady-state models explicitly account for conversion of HT to HTO in the environment and for dose from OBT as well as HTO, but until very recently, most models neglected one or more important pathways (Diabaté and Strack 1990). Models that have the necessary pathways, such as the twelve that were evaluated in the **Bio**sphere **M**odeling and **Ass**essment (BIOMASS) program of the International Atomic Energy Agency (IAEA 1996), are for the most part not published. NORMTRI (Raskob 1994) is a model that has all appropriate pathways, has been published, and did participate in BIOMASS. NORMTRI has many excellent features, but its HT pathways are not based on the latest experimental information on the behavior of HT in the environment (Davis et al. 1995; Davis and Bickel 2000). Another model that includes all essential pathways is that of Murphy (1986). This model, however, uses parameter values derived from observed data at the Savannah River Site and would be difficult to apply elsewhere. A third, NEWTRIT (Peterson and Davis 2002), has been coded at the request of the EPA into a version of the GENII code (Napier et al. 1988), GENII-NESHAPs. NEWTRIT calculates doses based on empirical ratios between compartments (e.g., concentrations in plant water compared with concentrations in air moisture) that have been selected to assure the conservatism needed for regulatory compliance. Thus NEWTRIT's predictions are perhaps too conservative for a realistic dose reconstruction. None of these models is probabilistic.

DCART was one of the twelve models evaluated in BIOMASS and was developed specifically to address realistically the conversion of HT to HTO in the environment and dose from OBT. The equations, parameter values, and assumptions in DCART are described in this report.

MODEL DESCRIPTION

DCART is a model that calculates doses (inhalation/skin absorption and ingestion of food and water) to adult, child (age 10 years) and infant (age 6 months to 1 year) from chronic releases of tritium gas (HT or T_2) and tritiated water (HTO) to the atmosphere. Driving input includes the HT or HTO source terms from each facility (in Ci/y) and dilution coefficients (χ/Q in s/m^3) calculated by a dispersion model. Wet deposition (HTO) and dry deposition (HT and HTO) to soil are both calculated as if they determined the concentration of tritium in soil water. However, the concentration in soil water, due to the difficulty of accounting for emission of HTO from soil, is actually determined by empirical ratios between air and soil water concentrations. Thus, the deposition pathway, although originally used by DCART, is effectively bypassed at present because empirical ratios are more robust than any attempt to model complex

processes simply, as befits DCART. Concentrations in precipitation, however, can be calculated with DCART, as can total deposition to soil before re-emission. For an HT release, HTO concentrations in air due to emission of HTO converted from deposited HT are estimated based on empirical ratios. The model calculates concentrations of tissue free-water tritium (TFWT or plant HTO) and OBT in edible plants (leafy vegetables, root vegetables, fruit or fruit vegetables (e.g., cucumber, tomato, eggplant, beans), grain, pasture, and hay) and animal products (milk, beef, pork, poultry and eggs). Doses are calculated using dose coefficients for HTO and OBT (ICRP 1996) and HT (ICRP 1995). Recommended parameter values and distributions of values for uncertainty analysis (see the Appendix) are specific to LLNL, to the extent possible. Dose from swimming in the LLNL pool is calculated. Concentrations of OBT in tree rings can also be calculated.

DCART has been tested within BIOMASS. Results of comparisons of predictions to observations, presented later in this paper, are among the best of all models participating. DCART has been evolving all the while, so the results presented for the BIOMASS evaluation may not be the same as would be obtained from the model described here, which is based on experience gained in BIOMASS. Predictions using the current version of DCART should be closer to observations and account for uncertainty as well.

Very few models, and certainly no regulatory models as yet, can predict doses from releases of HT and ingestion of OBT. Most models are restricted to releases of and doses from HTO. Of the models that include more than HTO, DCART should do as good a job of predicting doses from LLNL releases as any other model and do a better job than some, particularly for HT releases (to be discussed). Being developed in-house has many advantages. One is that the model is understood completely by the developer. Also, because DCART is a spreadsheet model, its calculations are more transparent than traditional compiled computer codes. Coupled with the risk assessment software, Crystal Ball®, DCART can produce probabilistic predictions as well as deterministic predictions. The model may be modified easily.

Equations in DCART

The default parameter values used in DCART are either specific to the Livermore site or similar sites or are the median of lognormal or mean of normal distributions of relevant observed data from the literature.

Deposition

Wet deposition of HTO is calculated:

$$\square \text{ (Bq m}^{-2}) = \square \text{ Q exp (-}\square \text{ x / u) } \square \text{T / (x u } \square\square) \qquad [1]$$

where:

$\square$ = washout coefficient (s^{-1}) (variable, depending on distance from source, stack height, and wind speed during rain (Belot 1998)); see Table A1.
Q = release rate (Bq s^{-1})
x = downwind distance in meters from the source
u = mean wind speed (m s^{-1}) for when it rains; sector, release height and year specific data are used when available; see Table A1.
$\square$T = duration of rainfall when plume is present (s); (calculated from fraction of time wind blows into a sector times fraction of time it rains times seconds in a year; see Table A1)
$\square\square$ = sector width (radians); 0.393

The value of $\square$ is varied to account for the fact that the washout coefficient is not constant throughout the plume, as in the case of aerosols, but depends on the shape of the vertical profile and hence on the distance from the source. DCART can calculate wet deposition at a location from all sources.

Then the concentration of HTO in soil water from wet deposition is calculated.

$$C_{sw,w} = (\square \text{ / (Precip + Irrig)) (0.001 m}^3 \text{ / L)} \qquad [1a]$$

where:

$C_{sw,w}$ = HTO concentration in soil water after rain or irrigating (Bq L^{-1})
Precip = mean annual precipitation (m^3 m^{-2} or m); see Table A1
Irrig = mean annual contribution of irrigation water (m) (California default 0.61 m; (Brewer 2001)

The concentration of tritium in precipitation is the same as [1a] except that the contribution from irrigation is not included.

Dry deposition is calculated for both HT and HTO as follows:

$$Dep_d = C_a \, v_g \, 3.15 \, 10^7 \text{ s y}^{-1} \qquad [2]$$

where:

Dep_d = dry deposition (Bq m^{-2})
C_a = concentration of HT or HTO in air at the location of interest (Bq m^{-3}); see Table A2.
v_g = deposition velocity (m s^{-1})(default for HTO: 5.3 10^{-3}; default for HT: 2.6 10^{-4}; see Table A3)

The amount of dry deposited HTO gets mixed into the amount of water entering the soil (i.e., the precipitation plus irrigation). Thus, as in [1a], the concentration of HTO in soil water (Bq L^{-1}) before re-emission from dry deposition is:

$$C_{sw,d} = (Dep_d / (Precip + Irrig)) (0.001 m^3 / L) \quad [2a]$$

Following the recommendations of the BIOMASS Tritium Working Group (IAEA 2003), DCART is calibrated so that the concentration ratio of soil moisture concentration to air moisture concentration ratio is 0.3 for a release of HTO (Table A3). Fractions have been observed up to 0.5 (Fellows et al 1990), which is the value recommended by the IAEA (2003) for screening models. The effect of this calibration is, as mentioned, to bypass the deposition pathways completely.

For a release of HT, the soil water concentration is calibrated so that the ratio of HTO in soil (Bq L^{-1}) to that of HT in air (Bq m^{-3}) is 6.0 (Table A3), which is the median ratio observed over natural soil during the Chalk River experimental HT release of 1994 (Davis et al 1995; Davis and Bickel 2000).

The concentration in the soil water is therefore

$$C_{sw} = f_r (C_{sw,d} + C_{sw,w}) \quad [3]$$

where:

f_r = fraction retained in soil water (variable and determined by calibration)

The retained fraction of HTO from deposited HT should be larger than for deposited HTO because HT diffuses deeper in the soil than does HTO.

Concentrations of TFWT and OBT in plants

For both HTO and HT releases, the HTO concentrations in plant water of leafy vegetables and pasture are given by (Raney and Vaadia 1965; Murphy, 1984) as:

$$C_{pw} = 1/\alpha [R_H C_{a\_HTO} / H_a + (1 - R_H) C_{sw}] \quad [4]$$

where:

C_{pw} = concentration of tritium in the plant water (Bq L^{-1} or Bq kg^{-1})
α = ratio of vapor pressure between HTO and H_2O (0.909)
R_H = relative humidity (LLNL annual mean default 0.69; Table A4)
C_{a_HTO} = concentration of HTO in air (Bq m^{-3})
H_a = absolute humidity (kg m^{-3})(LLNL annual mean default 0.0079); Table A4.
C_{sw} = concentration of tritium in soil moisture (Bq L^{-1})

Concentration of TFWT in Bq kg^{-1} fresh weight (fw) is obtained by multiplying C_{pw} by the fraction of the fresh weight plant that is water (1 – dry mass fraction; Table A4). Equation [4] is used to predict TFWT concentrations in most tritium research models (IAEA 2003), although it routinely appears to overestimate

TFWT concentrations by about 20% when compared with observations (IAEA 2003).

For both HTO and HT releases, concentration of OBT in all plants in Bq L^{-1} water equivalent (i.e., the amount of water produced when the dry matter is combusted; L kg^{-1}) equals the concentration in plant water (as calculated for leafy vegetables and pasture, eqn [4]) reduced by a discrimination factor that arises from isotopic effects in OBT formation. This isotopic discrimination results in a lower value for the specific activity (T/H - tritium atom/hydrogen atom) in the water of combustion compared with the specific activity in the plant water (McFarlane, 1976; Garland and Ameen, 1979). An isotopic discrimination factor of 0.7 has been chosen (Kim and Baumgärtner 1994). The concentration of OBT in kilograms fresh weight plant is obtained from:

$$C_{veg,OBT} = ID_p \, C_{pw} \, M_D \, W_{eq} \qquad [5]$$

where:

$C_{veg,OBT}$ = concentration of OBT in 1 kg of fresh weight plant
ID_p = isotopic discrimination factor for plants (0.7); Table A4
M_D = dry mass per kg fresh weight of plant material; Table A4
W_{eq} = water equivalent of dry plant matter (L kg^{-1}); Table A4

Concentrations of HTO in fruits, fruit vegetables and grain are calculated assuming 60% of the water in the fruit or fruit vegetable comes from air moisture and the other 40% comes from soil water (Davis 2003; Dinner et al. 1980; Fellows et al 1990; Table A4).

$$C_{fv,HTO} \text{ (Bq } L^{-1}) = (0.6 \, C_{a\_HTO}/H_a) + (0.4 \, C_{sw}) \qquad [6]$$

OBT in fruit, fruit vegetables and grain is calculated just as OBT in other vegetables (see eqn [5]), assuming that OBT is the product of photosynthesis and is translocated from leaves to fruit.

Concentrations of HTO in below-ground plant products (e.g., potatoes and root crops) are calculated as they are in NORMTRI (Raskob, 1994). This assumes that the water in the root crop is nearly in equilibrium with the soil water (Davis 2003; Table A4).

$$C_{rc,HTO} = 0.95 \, C_{sw} \qquad [7]$$

where:

$C_{rc,\,HTO}$ = the HTO concentration in root crops in Bg L^{-1}

OBT in root crops is calculated just as OBT in other vegetables (see eqn [5]), assuming that OBT is the product of photosynthesis and is translocated from leaves to roots.

Default parameters for relative humidity are the averaged annual values for the Livermore site for 1999 and 2001 - 2003[2] or year-specific. Default parameters for absolute humidity are derived from the mean tritium monitoring data (pCi/m^3 / $pCi/L = L/m^3$) for 1974 through 2003 or are year-specific[3]. Default parameters for fresh and dry weight fractions and water equivalent are mean values from Geigy (1981) (Table A4).

Resuspension/emission of HTO

The contribution of resuspended HTO to the HTO concentration in air for an HTO release is not modeled in DCART because resuspended HTO's contribution to air concentrations is insignificant except within 500 m of an elevated source (IAEA 2003). For a ground level release, reemission is not an issue if the model does not deplete the plume in the first place, as, for example, CAP88-PC does not for gases. However, because it is necessary to estimate HTO concentrations in air after a release of HT, emission is modeled for deposited HT that has been converted to HTO. At present, the HTO concentration in air moisture that is being inhaled or absorbed through the skin after a release of HT is calibrated to an observed ratio of 4.0 Bq L^{-1} HTO / Bq m^{-3} HT from the Chalk River HT release (Davis and Bickel 2000). The concentration of HTO in air moisture from a release of HT gas at 20 cm (plant height) is calibrated to a ratio of 6.0 Bq L^{-1} HTO / Bq m^{-3} HT observed during the Chalk River release (Davis and Bickel 2000; Table A4).

Concentrations of HTO and OBT in animal products

Data that describe the transfer of tritium from the environment to animals are minimal, but, for equilibrium conditions, the best evidence (Evans 1969; Moghissi et al. 1987) indicates that the tritium to hydrogen ratio in animal water and organic matter is the same as the environment to which the animal is exposed. Although tritium is too mobile to truly equilibrate with the environment, it is not unreasonable to develop a model for concentrations of tritium in animal products that assures that specific activity is maintained. This has been done for NEWTRIT (Peterson and Davis 2002) and for NORMTRI (Raskob 1994). The approach used for DCART is the same as that in NEWTRIT, except that DCART calculates the fractions of water contributed from plant water, plant organic matter, drinking water, and inhalation and skin-absorption based on user-provided diets rather than default diets, as in NEWTRIT.

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[2] Fifteen-minute data for all meteorological parameters are available for 1997 to the present at http://www.metdat.llnl.gov. An exceptionally high number of 15-minute readings of relative humidity were greater than 94% for the years 1988, 1998, and 2000, so the relative humidity for these years is not reliable.

[3] The decision to use air tritium monitoring data to estimate absolute humidity for DCART was based primarily on its having the longest time series and less uncertainty than using the absolute humidity calculated from temperature and relative humidity from the LLNL Meteorological Station.

The specific activity model followed in DCART is contained in the following calculation that gives the concentration of either HTO in water or OBT or water equivalent (Bq L^{-1} or kg^{-1}).

$$C_{aw} = \sum_{g,h,p}(C_{pw}\ (F_{pw} / T_w)) + \sum_{g,h,p}(C_{pweq}\ (F_{pweq} / T_w)) + C_a\ (F_{inh,s} / T_w) + C_{adw}\ (F_{dw} / T_w) \quad [8]$$

where:

C_{aw} = tritium concentration in animal water (HTO) or water equivalent (OBT) (Bq L^{-1})

$\sum_{g,h,p}$ = summation of the contributions of grain, hay or pasture to diet

C_{pw} = concentration of HTO in the water fraction of the plant ingested (Bq L^{-1}) (see eqn [4])

F_{pw} = kg of total water in the daily diet obtained from grain, hay or pasture (kg kg^{-1}); kg d^{-1} food type x times fresh weight fraction for food type x

T_w = total water ingested daily by the animal from all sources. This includes the water directly available from all foods, the water available after digested foods, the water obtained through skin absorption and breathing, and the water obtained from drinking water

C_{pweq} = concentration (Bq L^{-1}) in the water produced from foods after organic matter has been digested.

F_{pweq} = kg of water obtained daily after digestion of organic matter (kg kg^{-1}) in grain, hay or pasture; kg d^{-1} food type x times dry matter content of food type x times water equivalent factor of food type x

$F_{inh,s}$ = amount of water in L obtained from inhalation and skin absorption each day

C_{adw} = concentration of tritium in the animal's drinking water (Bq L^{-1})

F_{dw} = kg of water obtained daily from drinking water

As for the various plants, HTO and OBT concentrations for animal products (milk, beef, pork, poultry, and eggs, as eaten) are calculated separately. To obtain the concentration of HTO in Bq kg^{-1} of animal product, each product of eqn [8] (C_{aw}) is multiplied by the fresh water fraction (1 minus the dry matter fraction) of the particular animal product (Table A5). Similarly, to obtain the concentration of OBT in Bq kg^{-1} of animal product, each C_{aw} must be multiplied by the dry matter fraction times the water equivalent factor (Table A5) for the particular animal product.

Default parameters for fraction dry weight and water equivalent are mean values from Geigy (1981). Other parameter values for these equations (Table A5) have been obtained primarily from National Academy of Sciences data on animal nutrition (NAS 1994, 1996, 1998, 2001) supplemented with information from university agricultural and veterinary schools obtained from the World Wide Web (e.g., Ely and Guthrie 2001). Diets should reflect what is consumed just prior to slaughter. In the case of cows, the diet is based on what is known about the milk production (24.1 L d^{-1}) of the average California dairy cow (UCD 1998). The annual diet of dairy and beef cows (Table A5) is averaged over the

diet obtained from fresh pasture (four months of the year) and the diet of hay and grain for the rest of the year. Concentrations in animal products are quite insensitive to the type of feed ingested, but they are sensitive to the quantity of water ingested by each type of animal. Modern dairy cows need to drink large amounts of water to support modern (increased) milk productivity, and this will reduce the concentrations in milk by a few percent in DCART, because drinking water concentrations are assumed less than concentrations in plants (see next section).

Recently, a model for tritium concentrations in animal products based on hydrogen metabolism has been developed (Galeriu et al. 2001). This model accounts separately for each possible transfer from plants to animals. As mentioned, there is a paucity of data about the transfer of tritium to animals. The data that are available, however, indicate that some transfers occur preferentially when the system is not in equilibrium. These observations are supported by knowledge of hydrogen metabolism. For example, the bound hydrogen in the organic matter of plants that is digested to carbohydrates, proteins, and lipids by the animal is more likely to be synthesized into the organic matter of the animal than is the tritium atom that enters the body as water. The likelihood of transfers from diets to animals in decreasing order of occurrence is

- hydrogen in water to hydrogen in water
- hydrogen bound in organic matter to hydrogen bound in organic matter
- hydrogen bound in organic matter to unbound hydrogen in water
- unbound hydrogen in water to bound hydrogen in organic matter

A model like this is quite elegant and helps improve our understanding of the processes involved. Furthermore, it is finely tuned to all aspects of the animal, from the diet to the temperature of the ambient air to the activity level of the animal. However, the specific activity model may be preferred for simplicity and robustness. A comparison of the results of the DCART animal model with those of the Galeriu model is presented below.

Concentration of HTO in drinking water

If small surface water bodies are the source of the drinking water, as they might be for animals, the concentration of tritium in drinking water should be about that of soil moisture. Concentrations of tritium in small bodies of water are expected to have come from the atmosphere, just as soil tritium does, from an atmospheric release. Deposition rates of HTO to water and soils are reasonably similar. Thus concentrations in soil water and ponds may be similar. Exchange between soil water and pond water, although unlikely, would only serve to bring the concentrations closer together. For a screening level model (Peterson and Davis 2002), an assumption that the drinking water for animals is 50% that of air moisture is considered conservative. This assumption is similar to that of a screening level model assuming soil water at 50% air moisture (IAEA 2003) but is additionally based on observed tritium concentrations in small bodies of water relative to that in air moisture. More realistically, soil water concentrations are found to have about 30% the concentration of air moisture (IAEA 2003), so,

similarly, the concentration of surface drinking water concentrations for animals could be 30% of air moisture for a release of HTO. However, because atmospheric HTO depositing on the water surface is diluted by mixing with the mass of water, a water concentration of 30% or higher than that of air moisture is likely to be found only in very shallow bodies of water. The LLNL swimming pool could serve as a surrogate for a local small body of water. Since 1986, the concentration of water in the pool has been sampled, and air tritium concentrations have been sampled adjacent to the pool since mid-1990. As seen in Table 2, annually (e.g., mean of all pool samples divided by the mean of all air tritium samples), the pool water/air moisture ratio approaches 30%. The overall mean and median are about 16%, which probably represents the dynamics of tritium behavior in the Livermore Valley, given that releases are variable, have been dropping over time, and that loss of tritium can occur from bodies of water when wind direction changes, etc.

Table 2. Ratios of tritium in pool water (PW) to tritium in air moisture (AT) for corresponding time periods of sampling between 1990 and 2000. Only data greater than the lower limit of detection were used.

Year	Mean PW/Mean AT	Median PW/ Median AT	Number of samples
1991	0.156	0.281	11
1992	0.248	0.272	11
1993	0.102	0.132	7
1994	0.138	0.128	12
1995	0.151	0.231	8
1996	0.0776	0.0969	9
1997	0.143	0.148	6
1998	0.137	0.193	4
1999	0.102	0.163	3
2000	0.150	0.200	2
	Overall mean	Overall median	All ratios
All data '90 – '00	0.166	0.162	79

For a release of HT, the soil concentrations (HTO) predicted by DCART are the same as those in air moisture due to the conversion of HT to HTO in the soil. It is very unrealistic, however, to assume that, for an HT release, the concentrations in a small pond will equal those of soil water, because HT deposited on water is not converted to HTO as it is when it lands on soil. The pond concentration/HTO in air moisture ratio would be about the same after a release of HT as after a release of HTO, because most pond tritium comes from air HTO (in this case, emitted from the soil before being deposited on the water) even for an HT release. For this reason, in DCART it has been assumed, presumably conservatively, that surface water concentrations after an HT release

will be 30% that of the HTO in air moisture arising from conversion of HT to HTO in soil, just as estimated for a release of HTO.

When groundwater is the source of drinking water, many models assume that the concentration of drinking water is 1% that of air moisture (Moore et al. 1979; Till et al. 1981; USEPA 1989). This is a peculiar assumption because groundwater is unlikely to be contaminated from atmospheric releases except over the very long term. In addition, over the years at LLNL, as well as other places, the air concentration has varied by more than a factor of one hundred. Accordingly, in certain models, the estimated concentration in drinking water must also have varied by more than a factor of 100 over time. This illustrates the lack of viability in the assumption of drinking water being 1% the concentration of air moisture. Drinking water for people in the Livermore Valley is either from groundwater or surface water from distant sources, both of which would be not contaminated with locally produced tritium. Thus, in DCART it has been assumed that all drinking water for people is uncontaminated by LLNL tritium.

Doses

Dose from inhalation and skin absorption of HTO is calculated:

$$Dose_{inh\_HTO} = g_a \, C_{a\_HTO} \, I_{inh} * 1.5 \, DCF_{inh\_HTO} \quad [9a]$$

where:

g_a	= fraction of the consumed air arising from the contaminated source (assumed, at least initially, to be 1))
I_{inh}	= inhalation rate of adult, child or infant in $m^3 \, y^{-1}$ (Table A6)
1.5	= the factor that includes the dose from water intake through skin absorption relative to inhalation rate
$DCF_{inh—HTO}$	= ICRP (1996) dose conversion factor for inhalation of HTO for adult, child, or infant (Sv Bq^{-1}; Table A7)

Dose from inhalation of HT is calculated:

$$Dose_{inh\_HT} = g_{a\_HT} \, C_{a\_HT} \, I_{inh} \, DCF_{inh\_HT} \quad [9b]$$

where:

DCF_{inh_HT}	= ICRP (1995) dose coefficient for inhalation of HT for adult, child, or infant (Sv Bq^{-1}; Table A7)

In addition to direct inhalation of HT or HTO, there is also inhalation of HTO from HT that has been converted to HTO in the soil and emitted back to the atmosphere. The concentration of HTO from a release of HT at head-height is 4 (Bq L^{-1} HTO / Bq m^{-3} HT) (Davis and Bickel 2000; Table A6). Dose from HTO converted from HT is, of course, calculated as in eqn 9a.

Dose from drinking water is calculated:

$$Dose_{dw} = g_{dw} C_{dw} I_{dw} DCF_{HTO} \quad [10]$$

where:

g_{dw} =the fraction contaminated (0 for the Livermore Valley)
I_{dw} = drinking water consumed by adult, child or infant (L y^{-1}; Table A6)
DCF_{HTO}=the ICRP (1996) dose conversion factor for ingestion of HTO for adult, child, or infant (Sv Bq^{-1}; Table A7).

Ingestion doses are calculated:

$$Dose_{ing} = (\Sigma(g_f I_f C_{f,HTO}) DCF_{HTO}) + (\Sigma(g_f I_f C_{f,OBT}) DCF_{OBT}) \quad [11]$$

where:

g_f = fraction of the consumed food arising from the contaminated source (assumed, at least initially, to be 1)
I_f = consumption rate of the appropriate foodstuff (kg fw a^{-1}) by adult, child or infant (Table A6)
C_f = concentration of HTO or OBT in the appropriate foodstuff (leafy vegetables, fruit or fruit vegetables, root crops, grain, milk, beef, pork, poultry, and eggs) (Bq kg^{-1} fw)
DCF_{OBT}= the ICRP (1996) dose conversion factor for ingestion of OBT for adult, child and infant (Sv Bq^{-1}; Table A7)

Default parameter values are based on diets based on intakes in g per kg-day for different age groups as presented in the Exposure Factors Handbook (EFH) of the EPA (1999). Each diet contains about 80% of the mean caloric intake of an average adult, child age 10, and infant age 6 months to 1 year, respectively. The diet does not account for fish, nuts, and essential oils, which will not be affected to any degree by tritium releases in the Livermore Valley. The diet is thus relatively complete and, if it were assumed entirely contaminated with tritium, it should over-compensate for variations in the actual hypothetical diet of the Site-Wide Maximally Exposed Individual (SW-MEI). Default water consumption rates are intake rates for tap water from the EFH; inhalation rates are also from the EFH; Table A6).

Dose coefficients are (ICRP 1995, 1996) are summarized in Table A7.

In addition to the major contributors to dose (i.e., inhalation and ingestion), there is another pathway at LLNL to dose – swimming in the LLNL pool. The contribution from this is negligible compared with doses from inhalation and ingestion. Nevertheless, it would be wrong to omit the calculation of the swimming dose from DCART.

Dose from immersion in water (Osborne 1968) can be expressed as:

$$Dose_{imm} = C_p A_{bs} I_s H_s T DCF_{HTO} \quad [12]$$

where:

C_p = concentration of HTO in pool water (Bq/g) (observed or computed from known air concentrations)
A_{bs} = surface area of the body (m^2); see Table A9.
I_s = intake rate of HTO through skin (5.1 L/min m^2)
H_s = humidity at skin surface (0.02724 g per L at mean temperature (28° C) of LLNL pool)
T = duration of time swimming in minutes
DCF_{HTO}= the ICRP (1996) dose conversion factor for skin absorption for adult, child and infant (Sv Bq^{-1}; Table A7)

For some of the years when the pool water was not sampled for tritium, pool water concentrations can nevertheless be estimated. Air moisture concentrations measured at the LLNL Discovery Center (previously the Visitors Center) are quite well correlated with air moisture concentrations measured adjacent to the pool. Pool water concentrations are in turn correlated with air moisture concentrations adjacent to the pool (Table 2). Thus, although with increased uncertainty, the annual mean air moisture concentrations measured at the Discovery Center can be used to estimate pool water concentrations.

Seasonal vs annual mean concentrations in vegetables

Regulatory models calculate doses based on annual average releases and annual average concentrations in foodstuffs and in air, because the calculations are relatively easy to do and err on the side of conservatism. Food, either for animals or people, that is not eaten fresh, is assumed stored and eaten later. Radiological decay accounts for loss of activity during this time. DCART, like regulatory models, assumes that the vegetables and animal products contain the annual average concentrations when they are ingested but does not account for radiological decay because of the 12.32 year half-life of tritium and the assumption that no foodstuff is stored for more than eight months and most foods are eaten within days of harvest.

In the Livermore Valley, vegetables can grow nearly year round, although certain types of vegetables may only grow in particular seasons. In summer, if things are to grow, they need to be watered. To account for drought conditions, an irrigation pathway was added to DCART. However, as DCART evolved, the ratios of tritium in soil moisture to either HTO in air moisture for an HTO release or HT in air for an HT release were set to experimentally observed values. Because of this calibration, soil tritium concentrations have to be the same regardless of water content of the soil or the source of the soil water. The irrigation pathway is therefore unnecessary unless the irrigation water were to contain tritium from LLNL operations. As the water content of the soil decreases, in order to maintain the set relationship between soil and air concentrations, the fraction of HT or HTO retained after dry deposition drops. DCART (eqn [1]) needs only about 8 cm of water input to the soil annually to keep the fraction of HTO retained by the soil at a non-negative value. This problem is academic, because edible vegetation needs more than 15 cm of water per year to grow, and

because more than 15 cm of rain has fallen every year since the laboratory opened.

In DCART, any differences between predicted winter and summer concentrations in vegetation are due to different assumptions about absolute and relative humidity and different wind speeds and direction. Summer absolute humidity is higher and summer relative humidity is lower than during the winter, but obviously the annual average accounts for both, and the use of mean concentrations in vegetables will account for differences in concentrations depending on the season. The uncertainty is such that it would be pointless to attempt to model winter vegetables differently from summer vegetables, even though some vegetables only grow well at certain times of year. In addition, if predictions were to be made seasonally, a seasonally adjusted wind rose would have to be used in place of the annual wind rose normally prepared for dispersion models.

The above discussion of seasonal changes in absolute and relative humidity refers only to a release of HTO. For an HT release, the concentrations in vegetation are independent of absolute and relative humidity because the HTO in air moisture is set equal to the observed ratio of 6 Bq L^{-1} in air moisture to Bq m^{-3} HT in air at plant height (Davis and Bickel 2000; Table C4). Thus, the only seasonal effect on tritium concentrations at a particular location would be due to differences in wind patterns.

UNCERTAINTY AND SENSITIVITY ANALYSIS OF DCART

All uncertain parameters that are not source, receptor or time specific have been given distributions (see the Appendix) to be sampled either by Monte Carlo (MC) techniques or Latin Hypercube Sampling (LHS) using Crystal Ball® to assess the 95 percent confidence interval on doses to adult, child, and infant and to determine the parameters to which the model is sensitive. Obviously, the uncertainty about the input that drives the model (source term, χ/Q estimated from the dispersion model, or the average annual observed air concentrations at a particular location) will have a major effect on the uncertainty on the doses. As well, these same parameters, when varied, may be those to which the model is most sensitive. Thus, the results of an actual uncertainty and sensitivity analysis must be unique to each annual input at LLNL, given that source locations, the magnitude of the sources, and the meteorological parameters may vary considerably each year. An assessment of the uncertainty and sensitivity for each year of the LLNL dose reconstruction will be calculated and reported when the dose reconstruction is carried out. For now, the sensitivity and uncertainty analyses presented here apply only to the parameters in the Appendix and to the specific distributions assigned to them. The results shown below were calculated by Crystal Ball® assuming an input of unit air concentration (1 Bq/m^3 of HT or HTO) and running DCART 10,000 times with LHS. Because the precipitation pathway is bypassed in DCART and calculations are driven by concentrations of tritium in air rather than by source terms, only those parameters listed in Tables

A3 – A7 are included in the analysis. The analysis carried out in this way will be referred to as the generic analysis.

Uncertainty analysis

Assuming input of a known air concentration, the magnitude of the 95 percent confidence interval when all generic parameters are varied is shown in Table 3. In addition, the ratio between the mean of the output distribution and the deterministic dose is shown for all generic parameters.

Table 3. Magnitude of 95 percent confidence intervals and comparison between means of the probabilistic and deterministic outputs predicted by DCART for unit air concentrations of HT and HTO (simulating releases of HT and HTO, respectively). Results are shown for releases of HT and HTO when all generic parameters are varied, when all are varied except the dose coefficients (DCF), and when all are varied except distributions on diet.

	97.5 / 2.5 percentile dose			Mean of probabilistic output / deterministic output		
HT	All	Less DCF	Less diet	All	Less DCF	Less diet
Adult	6.8	6.1	4.0	2.6	1.2	2.4
Child	6.3	4.9	4.5	2.3	1.2	2.2
Infant	12.	9.8	4.8	2.3	1.2	2.3
HTO						
Adult	4.4	3.7	2.4	2.4	1.1	2.3
Child	4.1	2.9	3.0	2.2	1.1	2.1
Infant	7.7	6.4	3.2	2.2	1.1	2.2

The uncertainty is greater on doses from HT releases than on doses from releases of HTO because of the large uncertainty on the conversion of HT to HTO in the environment (see Tables A3 and A4). Uncertainty is highest for infant doses, in large part due to uncertainty in infant diet. The magnitude of the 95 percent confidence interval is reduced significantly when either the uncertainty on the dose coefficients or the uncertainty on diet is removed. If uncertainty on both the dose coefficients and diet is removed, the magnitude of the 95 percent confidence limit for a dose from a release of HT is reduced to a factor of 3.5; for a release of HTO, the overall uncertainty is less than a factor of 2.

Removing the uncertainty on the dose coefficients not only reduces the overall uncertainty on all doses considerably, but also brings the mean of the probabilistic doses close to that of the deterministic doses. The difference of more than a factor of two between the probabilistic and deterministic doses is due to the fact that distributions of dose coefficients are skewed very high compared with recommended ICRP values. Yet, until the ICRP or other regulatory body agrees to higher deterministic values for a dose coefficient, the values currently recommended must be used.

As mentioned, more uncertainty will be introduced when the distributions on source terms, and dilution factors are included.

Although deposition pathways in DCART are bypassed, the precipitation sub-model can be used by itself to estimate concentrations of tritium in rainfall. In some circumstances, knowing the confidence that can be placed in the predictions of concentrations in rainfall is important.

To test uncertainty associated with predicted concentrations of tritium in rain, relatively high, hypothetical release rates from both stacks and area sources were chosen (2000 Ci from the Tritium Facility, 4.4 Ci from the B612 yard and 7.3 Ci from the B331 waste accumulation area (WAA)). Meteorological data (Table A1) were those for LLNL for 2002. For the precipitation sub-model, rainfall concentrations must be calculated at a particular location, so tritium concentrations in rain from each source were calculated at the Discovery Center, which is adjacent to the location of LLNL's historical site-wide maximally exposed individual. The parameters that were sampled are those from Table A1. Uncertainty on rainfall concentrations is high, ranging from a ratio (97.5/2.5 percentile) of 7.7 for rain containing tritium from the B331 WAA to 10.7 for rain containing tritium from the B331 Stacks. LLNL has monitored concentrations in rain for many years at the Discovery Center and elsewhere. At some point, the precipitation sub-model can be tested with the LLNL data.

Sensitivity analysis

The sensitivity of predictions to specific parameters strongly depends on the set of input parameters and the endpoint. The input to DCART for this sensitivity analysis, as for the uncertainty analysis above, was unit concentration of HT or HTO in air (Bq m^{-3}). The endpoint for this analysis was the total dose to adult, child and infant from HT or HTO concentrations in air, which directly influence dose at the location of the exposed individual. As with the uncertainty analysis, only the parameters listed in Tables A3 to A7 have been included in the analysis.

The results of this generic sensitivity analysis are shown in Table 4. The four most important parameters to each dose endpoint, as determined by the rank correlation coefficient, are shown.

All of the parameters in Table 4 have relatively large uncertainties that are either due to lack of knowledge, to natural variability, or to a combination of both. The negative correlation coefficient for leafy vegetable intake (I_l) is due to the fact that I_l is correlated with fruit vegetable intake (I_f; Table A4). The dose is more sensitive to I_f than to I_l, so if I_l increases, then I_f decreases, as does the dose.

Table 4. Four most significant parameters and their rank correlation coefficients for doses to adult, child, and infant derived from unit air concentrations of HT and HTO in DCART.

Parameter	Dose from HT	Parameter	Dose from HTO
Adult		Adult	
HTO in plant (Bq L^{-1}) / HT in air (Bq m^{-3})	0.49	Fruit or fruit vegetable intake	0.52
Fruit or fruit vegetable intake	0.49	HTO dose coefficient	0.38
Leafy vegetable intake	-0.32	Absolute humidity	-0.35
HTO in soil (Bq L^{-1}) / HT in air (Bq m^{-3})	0.31	Leafy vegetable intake	-0.34
Child		Child	
HTO in plant (Bq L^{-1}) / HT in air (Bq m^{-3})	0.54	HTO dose coefficient	0.50
HTO dose coefficient	0.35	Absolute humidity	-0.36
Fruit or fruit vegetable intake	0.34	Fruit or fruit vegetable intake	0.35
HTO in soil (Bq L^{-1}) / HT in air (Bq m^{-3})	0.31	OBT dose coefficient	0.34
Infant		Infant	
Milk intake	0.50	Milk intake	0.57
HTO in plant (Bq L^{-1}) / HT in air (Bq m^{-3})	0.44	Fruit or fruit vegetable intake	0.39
Fruit or fruit vegetable intake	0.40	HTO dose coefficient	0.37
HTO dose coefficient	0.28	Leafy vegetable intake	-0.29

When a prediction is sensitive to a parameter, the parameter should be examined to see if its uncertainty can be reduced. In the case of the parameters in Table 4, nothing at this point in time can reduce the size of the distributions (Tables A4, A6, and A7). More experiments could refine the uncertainty on the ratio between HTO in plant water and HT in air, but the predictions of dose from an HT release would remain sensitive to the ratio. When the ICRP or other regulatory body addresses the uncertainty in the dose coefficients, sensitivity to the dose coefficients should be reduced. The uncertainties in ingestion rates are due to natural variability and presumably cannot be reduced. Sensitivity of the model to some parameters will always exist, regardless of how well the values are known and described.

A sensitivity analysis was also run on the precipitation sub-model. The model is very sensitive (correlation coefficients between 0.57 and 0.75 depending upon the source of tritium) to the washout coefficient and to the fraction of time the wind blows into a sector when raining.

TESTING DCART

BIOMASS scenarios (concentrations of tritium in plants, soil, and rain)

Over the past nearly twenty years, several international efforts have been directed at testing models that calculate doses from releases of various radionuclides to the environment. The most recent of these efforts has been the International Atomic Energy Agency's BIOMASS (BIOspheric Modeling and ASSessment) program (IAEA 2003). One of the groups, the Tritium Working Group, analyzed the results of the following five scenarios.

- Scenario 1: Modeling of the steady-state behavior of HT and HTO in the environment when atmospheric releases are assumed to be on average nearly constant and a steady-state equilibrium has been reached.
- Scenario 2: Model-model intercomparison exercise for predicting the rise of tritium from contaminated ground waters.
- Scenario 3: A test of chronic atmospheric release models using Canadian data.
- Scenario 4: A test of chronic atmospheric release models using Russian data.
- Scenario 5: A test of chronic atmospheric release models using French data.

DCART has been tested in all scenarios except Scenario 2. DCART was under development during the period of the BIOMASS program, and the current version will give somewhat different results. Differences between the version tested and the current version are mentioned below. For Scenarios 1, 4 and 5, air concentrations used as input to DCART had to be predicted by dispersion models. Two different models were used for BIOMASS depending upon the form of the meteorological input data. One was the dispersion model from CAP88-PC (Parks 1997) and the other was the Canadian Standard N288.1 (CSA 1987). Results of these dispersion models will not be discussed here, except as they affect concentrations predicted by DCART.

Predictions for BIOMASS scenarios had to be submitted before any observations were released. This way, modelers could not tune their results. However, once the observations were revealed, modelers could revise their predictions based on having found mistakes in the codes or on having misunderstood the scenario description. The final results presented in the reports (IAEA 2003) are revisions, as are the results presented below. (If a participant chose not to revise obviously wrong predictions, they were omitted from the analysis.)

Those aspects of DCART's performance in BIOMASS that are relevant to using DCART to reconstruct doses from routine releases at LLNL are presented below.

Scenario 1 – Model Intercomparison

Scenario 1 assumed a continual release of a gram of either HT or HTO from a 60 m stack over the course of a year. Frequency of occurrence of stability classes and mean wind speeds for each stability class were provided, as were deposition velocities for HT and HTO. Endpoints asked for were tritium concentrations in air, soil water, plant water and combustion water (OBT) (all in Bq L^{-1}) at every 100 m, from 100 to 1000, and every 1,000 meters thereafter to 10,000 meters. The Canadian dispersion model (N288.1), with a vertical dispersion parameter recommended in the scenario description, was used for calculation of air concentrations.

Because this exercise was a model intercomparison, no answer could be right or wrong. Model intercomparisons simply reveal the degree of consensus (or lack thereof) between modelers. Eleven models participated in the HTO scenario, while nine participated in the HT scenario. LLNL joined BIOMASS after this scenario closed, but the scenario was used as the first test of DCART early in its development. Among all models tested, the variability in model results was higher close to the source due to the use of different dispersion assumptions. For example, some modelers included the contribution from resuspended HTO in their estimates of air concentrations. Agreement was better at distance. Because LLNL's major interest in a dose reconstruction will be dose to an individual living offsite, results between the participating models and DCART are compared in Table 5 for 1,000 and 5,000 m from the source. For the release of HTO, results have been normalized to the median air concentration of each data set so that, in effect, all models are starting with the same air concentration, thus nullifying the effects of dispersion modeling. For the HT release, HT concentrations in air cannot be normalized because this endpoint was not requested, so the effects of dispersion modeling carry through the other predictions. The estimation of HTO in air after a release of HT is different for each model.

The modelers agree quite well on how to model HTO, because the spread in predictions is less than a factor of 2.5, except for soil, which has a spread greater than a factor of 5.9. DCART's results are very close to the median concentration predicted by the other modelers, except for soil. DCART's soil water/air moisture ratio was 0.44 (now calibrated at 0.30), which is low compared with other models in this scenario, which were using higher, screening levels (IAEA 2003). High soil water/air moisture ratios are due to high deposition velocities coupled with explicit or implicit low emission rates. In general, soil pathways are not considered very important to a tritium dose model.

Table 5. DCART predictions of HTO in air, plant water, and soil, and OBT in plants at 1,000 and 5,000 m from the 60 m source of HTO or HT divided by the median, maximum and minimum predicted concentrations submitted by participants in Scenario 1 of BIOMASS. Results from the HTO release have been normalized to the same starting air concentrations.

	DCART median		DCART maximum		DCART minimum	
Ratios DCART/other models - continuous release of one g HTO over a year						
	1000 m	5000 m	1000 m	5000 m	1000 m	5000 m
HTO in air	[1.0]	[1.0]	[1.0]	[1.0]	[1.0]	[1.0]
Plant water	1.0	0.99	0.81	0.78	1.2	1.2
OBT in plants	0.96	0.95	0.66	0.66	1.7	1.3
HTO in soil	0.73	0.68	0.39	0.34	2.3	2.3
Ratios DCART/other models - continuous release of one g HT over a year						
	1000 m	5000 m	1000 m	5000 m	1000 m	5000 m
HTO in air	2.9	2.2	1.0	1.0	15.	9.8
Plant water	1.2	1.0	0.52	0.53	12.	5.4
OBT in plants	1.4	1.1	0.42	0.16	12.	5.4
HTO in soil	0.93	0.90	0.33	0.28	12.	5.1

The uncertainty in the predictions of concentrations of HTO in the environment from a release of HT is much greater, ranging up to a factor of 36 between the lowest and highest predictions for soil concentrations at 1000 m. Between the time when the participants submitted results and when DCART's predictions were calculated, much had been learned about the concentrations of HTO in the environment after a release of HT because of ongoing analysis of the data from the 1994 HT release at Chalk River Laboratories (Davis and Bickel 2000). These studies revealed that more HTO is emitted than had previously been thought. Thus, as can be seen, DCART's prediction for HTO in air moisture is the highest (although this may be partially due to the results of dispersion modeling), and DCART's predictions for the other media are amongst the highest. The ratio of HTO in air moisture to HT in air for this comparison was 5.7 based on Chalk River observations. As mentioned, this ratio in DCART is now based on 6.0 (with uncertainty) due to improved understanding of uncertainties on the Chalk River data.

Scenario 3 – Chalk River Laboratories, Chalk River, Canada

Scenario 3 (IAEA 2003), based on observations at Chalk River Laboratories (CRL), Ontario, Canada, was the first opportunity to test DCART against field observations. Modelers were provided with comprehensive meteorological data measured at CRL for June 5 – August 10, 1995. These included hourly and daily averages of relative humidity, air, leaf and soil temperatures, rainfall and rain intensity, wind speed, wind direction and stability class. Source terms (Bq s^{-1}) were provided for the reactor stack, the reactor building and a contaminated lake. In addition, measured daily concentrations of tritium in air (Bq m^{-3}) were provided for about half the days of the study for three locations at CRL. Average observed air concentrations were used to drive DCART, as was the

intention of the scenario developer, to limit the uncertainty arising from the use of various dispersion models.

The observational data at three sampling locations against which the models were tested included:

1. TFWT sampled at 9:00 (representing nighttime processes) and 15:00 (representing daytime processes) daily,
2. Soil water concentrations sampled at 9:00 and 15:00 daily,
3. OBT in plants sampled on June 28, July 12 and August 9, and
4. Concentrations of tritium in rainfall at one site.

As well as calculating the above endpoints, plus a 24-hour mean concentration in plant water, modelers were asked to provide the 95% confidence intervals on all predictions.

DCART's predicted to observed (P/O) ratios for the chronic endpoints (Table 6) were similar to those of the other participants. With the exceptions of rain and OBT on August 9, DCART overestimates the observations by a small amount. If predictions were made again with the present DCART, P/O ratios would be closer to 1.0. The low P/O ratio for rain was due to the observed ratio (rain/air moisture in Bq L^{-1}) being about 0.8, which is higher than any model result. In fact, only DCART and one other model had uncertainty bounds that overlapped the uncertainty on the observed concentration in rain. As seen in Table 6, the P/O ratio for OBT in grass drops over time. This is because the observed OBT in plants increased over time relative to the air concentrations. This too is an unexpected result and was not predicted by any model.

The various ratios predicted by DCART varied depending upon the location and the time of day. The soil/air moisture ratio ranged between 0.34 and 0.44; the TFWT/air moisture ratio ranged from 0.85 to 1.0; and the rain/air moisture ratio ranged from 0.19 to 0.28; the OBT/TFWT ratio was 0.8. In the DCART for dose reconstruction, the soil/air ratio for HTO is 0.3 and the OBT/TFWT ratio is 0.7.

Table 6. Predicted to observed ratios for DCART for the endpoints of the CRL Scenario of BIOMASS.

Endpoint	Location 1	Location 2	Location 3
Rain, 24 hour	0.30		
Soil, morning	1.4	1.4	1.7
Grass, HTO, morning	1.1	1.1	1.1
Grass, HTO, afternoon	2.6	1.2	1.4
OBT, June 28	2.5	1.9	2.9
OBT, July 12	1.8	------	2.6
OBT, August 9	1.0	0.69	1.2

At the time, DCART could not be run with Monte Carlo uncertainty analysis, so parameter perturbation was used to assess uncertainty on the predictions. The median values of various parameters, plus or minus standard deviations, were calculated from a database of variables used in DCART. These were then combined as a set of values that would produce a low average predicted concentration and a set of values that would produce a high average predicted concentration. The three sites are very different, so the uncertainty on the predictions varied with the site. Nevertheless, in general, the uncertainty on the plant water concentrations and OBT was about a factor of 3.3 (high/low). Uncertainty in soil concentrations was roughly a factor of 50. Uncertainty was lowest for the 24-hour period and highest for the 6-hour daylight period, probably because more numbers contributed to the averages for the longer time period. Because these uncertainty estimates were extremes, the demonstrated uncertainty was higher than the 95% confidence interval that would have been predicted using Monte Carlo analysis.

In the current stochastic version of DCART, the ratios of the 97.5 percentile divided by the 2.5 percentile of the output distributions are about two, three and six for HTO in pasture, OBT in pasture, and soil water concentrations, respectively. Uncertainty in soil water concentrations in this version of DCART are driven by the assumption that the soil water to air moisture ratio is always 0.30.

Scenario 4 - Russian Federal Nuclear Center, Sarov, Russia

The Sarov Scenario (IAEA 2003) asked for concentrations in air, plant water, soil water, snow, and rain at 500, 1,700, 2,800, 3,000, 5,000 and 7,600 m from a source that annually emitted equal quantities of HT and HTO. The modelers were provided with annual emissions in arbitrary tritium units for each year after the start-up of operations. Emissions peaked in the 5th year of operations at 120 arbitrary tritium units; in the 10th year, 12 units were emitted; in the 15th year, 4 units were emitted; and for years 16-20 post start-up (1985-1989), 1 unit per year was emitted. The actual release rate from Sarov was kept secret this way, so all results that the modelers discussed were the normalized ratios of measured or predicted concentrations to the measured or assumed release rates.

Unfortunately, the only long-term averages that were monitored were for air HTO[4], so the only model predictions that could be compared meaningfully with observations were air concentrations based on assumptions about absolute humidity. Thus the Sarov scenario really only tested the dispersion model used (CSA-N288.1). Nevertheless, this scenario has been described here in detail because of one of the conclusions of the BIOMASS Tritium Working Group (IAEA 2003). At Sarov, there was some evidence for tritium retention in biosphere media after a significant decrease in emission rates (in this case, over 2 orders of magnitude during 20 years of releases). The observed tritium air concentrations showed a slight increase in time when normalized by the release

[4] Plant and soil water samples were collected on only one day each year, so model predictions could not be compared meaningfully with the observations.

rates. However, the magnitude of the increase was comparable to experimental uncertainties, which demonstrates a need for further study to determine if retention really does occur and, if necessary, to determine those processes responsible for it so that it can be taken into account in long-term assessment models. Two of the participating models attempted to account for memory effects, but results were not distinguishable from those of DCART or the other models.

The proportion of HTO contributed from the HT releases could not be measured, and therefore there was no discussion about the proportion calculated in the different models.

Scenario 5 – Centre Energie Atomique, Valduc, France

The Valduc Scenario (IAEA 2003) described three sources of tritium with annual releases of HTO and HT from 1983 to 1998. At specified locations, modelers were asked to predict annual average tritium concentrations in air, plant water, and rainwater. Six models participated. Concentrations of OBT in tree rings of birch for 1983 to 1988 and OBT concentrations in oak leaves were also endpoints. The dispersion modeling for this scenario was complex because of the three sources, uneven terrain and numerous receptor sites. CAP88-PC was used as the dispersion model, and it performed as expected (Peterson 2003[5]). Stations 1-4 ranged from 3500 to 6900 m from the sources. The distances from the sources to the birch tree ranged from 9000 to 9500 m.

Vegetation was sampled monthly at four locations for eleven years. For this comparison of modeling results, concentrations in vegetation were normalized based on observed air concentrations to remove the bias of different dispersion models. Normalized concentrations of vegetation are compared with observations in Table 7. DCART's predicted concentrations in TFWT lie for the most part midway in the range of the other modeling results. All participants overestimated TFWT concentrations.

Rainfall was collected under oil monthly for a time-integrated concentration of tritium in rain at the same locations for the same time (Table 8). Because DCART calculates concentrations in rain independently of any dispersion model (eqn [1]), there is no reason to normalize rain concentrations to observed concentrations of tritium in air. On average, DCART shows a small tendency to underestimate concentrations of tritium in rain (Table 8).

DCART's predictions of OBT in birch tree rings ranged from 0.82 to 3.2 of the observed values; DCART's average P/O ratio for the six years was 1.8. DCART generally overestimates the concentrations in tree rings, but it was on average closer to the observations than any other model (with overall average P/O ratios of 2.4 –3.3). It was impossible to normalize the predictions of concentrations of

[5] All predictions for the Valduc locations fell within a factor of three of the observations. Only about 37% of the predictions were higher than the observations, which is similar to CAP88-PC's performance at locations similarly distant from LLNL.

OBT in the beech tree rings because no air concentrations were measured at the locations of the trees. DCART's overestimation of OBT in tree rings is probably not due to an overestimation of HTO in air by CAP88-PC (see footnote 4).

Table 7. Predicted-to-observed (P/O) ratios for concentrations of TFWT in grass at four locations at Valduc. Predictions were normalized to air moisture concentrations at each location. DCART's normalized averages for 1988 through 1998 are compared with the range of normalized predictions submitted by the other participants.

Year	DCART's Normalized P/O Values			
	Station 1	Station 2	Station 3	Station 4
1988	3.4	2.6		2.3
1989	2.0	2.6		2.9
1990	1.0	2.2		1.7
1991	2.3	1.9		3.3
1992	1.2	1.8		1.5
1993	4.6	1.5		3.0
1994	1.4	1.6	2.1	1.8
1995	0.80	1.3	2.1	1.5
1996	2.7	1.9	2.0	1.8
1997	3.8	3.6	1.8	3.1
1998	1.3	1.7	1.6	2.2
DCART normalized average	2.2	2.1	1.9	2.3
Modeler's Range – normalized averages	1.7 – 2.6	1.8 – 3.3	1.7 – 2.6	1.8 – 2.9

Table 8. Predicted-to-observed (P/O) ratios for DCART and the range of the average P/O ratios for all models for tritium in rain at Valduc.

Year	DCART's Normalized P/O Values			
	Station 1	Station 2	Station 3	Station 4
1988	0.50	0.86		0.24
1989	0.72	1.2		0.57
1990	0.63	0.65		0.48
1991	1.9	1.3		1.3
1992	0.72	0.71		0.45
1993	1.0	0.84		0.52
1994	1.5	0.87	0.82	1.1
1995	0.69	0.84	0.56	0.62
1996	1.4	1.2	0.56	0.91
1997	0.78	0.99	0.37	0.49
1998	0.79	1.2	0.66	0.56
DCART average	0.97	0.97	0.59	0.66
Modeler's Range	0.12 – 2.2	0.32 – 3.9	0.39 – 1.4	0.18 – 1.5

Modelers also had to predict concentrations of OBT in oak leaves at 36 locations at the Valduc site. These locations ranged from 1.7 to 15 km from the vicinity of the tritium sources. P/O ratios of DCART ranged from 0.26 to 1.8, with a mean of 0.92 and a median of 0.83. The average results of the other five models ranged from 1.4 – 2.2. No measurements of tritium in air were taken at the location of the oak trees, so data could not be normalized to known air concentrations. DCART's small underestimation of observations is probably due to underestimated air concentrations at distance by CAP88-PC (Peterson 2003).

Average ratios and range of ratios of various endpoints to air moisture are shown in Table 7 for DCART. DCART's OBT/TFWT ratio for grass and oak leaves was 0.8; for beech tree rings it was 0.4 (Kalin et al. 1995).

Table 9. TFWT and rain concentrations predicted by DCART normalized to air moisture concentrations predicted by DCART for four locations at Valduc. The average and range of DCART results for 1988-1998 are presented.

	Station 1	Station 2	Station 3	Station 4
Average TFWT	1.0	1.3	1.0	1.1
Range TFWT	0.46 – 2.4	0.66 – 1.8	0.64 – 1.5	0.60 – 1.7
Average Rain	0.24	0.69	0.14	0.13
Range Rain	0.12 – 0.57	0.36 – 0.92	0.10 – 0.20	0.08 – 0.18

BIOMASS scenarios – summary

Out of four scenarios, only a small fraction of the data was appropriate to test chronic release models. Although there were many measurements of concentrations of HTO in air, these data are primarily only useful to test dispersion models. At Chalk River, TFWT samples were taken frequently enough to be averaged meaningfully; the monthly TFWT samples taken at Valduc may not be quite as representative of an annual average. Because it is expensive and time-consuming to sample OBT, oftentimes OBT measurements are too few to test models adequately. At Chalk River, it would have been better to have had more OBT measurements, but at least the Chalk River OBT measurements corresponded to tritium measurements in air. In contrast, at Valduc, although there were many OBT measurements, there were no corresponding air concentrations with which to compare them. Samples for model testing need to be collocated in time and space. At Valduc, samples of rainfall and air concentrations met this criterion, thus providing reliable ratios for model testing. In contrast, the rainfall data for Chalk River were not collected at the same location as any of the air samplers, although distances between them were not great.

Models for animal products compared

Experiments (Moghissi et al. 1987) and monitoring of equilibrated environments (Evans 1969) have shown that the T/H ratio in animals equals that of their

environment when all compartments of that environment have the same T/H ratio. The calculation of tritium concentrations in meat and eggs in DCART is based on these observations, as is the calculation in NORMTRI (Raskob 1994). The models in DCART and NORMTRI are very similar. Concentrations of OBT in beef and poultry are the same in both models; all other concentrations are about 10% higher than those of NORMTRI, except for HTO in eggs, which is about 20% higher. Thus DCART does not underestimate concentrations in animal products compared with NORMTRI, nor does it overestimate significantly.

Recently Galeriu et al. (2001) developed a model based on the hydrogen metabolism of animals. This model improves upon the type of regulatory model that uses a transfer factor (d kg^{-1}) to calculate concentrations in animals from intakes of feed and water. As mentioned, each possible transfer from diet HTO or OBT to animal HTO or OBT does not have an equal chance to occur. These transfer parameters vary depending upon various assumptions about the metabolic status of the animal. When diets are not uniformly contaminated, this approach potentially gives a better estimate of fractions of HTO or OBT in the animal and thus a better estimate of dose than does the specific activity model.

Concentrations in animal products have been compared between the Galeriu model and DCART assuming the same ingestion rates and concentrations of tritium in the diet. Results for all animal products comparing the deterministic predictions of the Galeriu model with the 95% confidence intervals of DCART are shown in Figure 1. All results from the Galeriu model fall within the uncertainty bounds of DCART. Thus, although the Galeriu model is potentially more accurate than DCART, its results are nevertheless encompassed by DCART's 95% confidence interval.

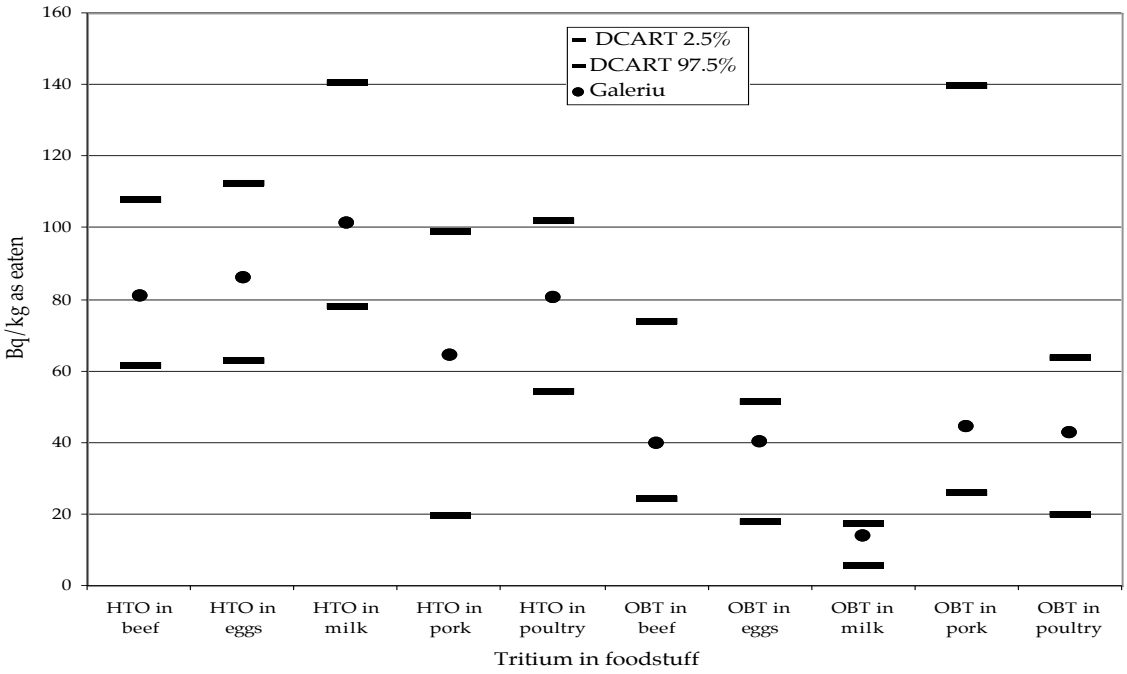

Figure 1. Deterministic concentrations of HTO and OBT in animal products predicted by the Galeriu metabolic model lie within the 95 percent confidence interval on concentrations predicted by DCART. Only parameters for animal intake have been varied in DCART, and all concentrations in pasture, hay, grain, and water were the same for both model calculations.

Doses compared: DCART vs NEWTRIT

DCART has also been compared with NEWTRIT, a relatively simple model geared to regulatory compliance (Peterson and Davis 2002). Doses will differ because of the different diets used in each model, so doses will not be compared here. Other differences stem from the greater scope of DCART (more detailed modeling) and the more realistic (vs. conservative) choice of parameter values, coupled with uncertainty analysis, in DCART.

The mean and the 2.5 and 97.5 percent confidence limits of DCART's stochastic predictions are compared graphically with NEWTRIT's deterministic prediction in Figure 2 (for HT) and Figure 3 (for HTO). Initial air concentrations were the same (1 Bq m^{-3}), as was the absolute humidity (8 g m^{-3}).

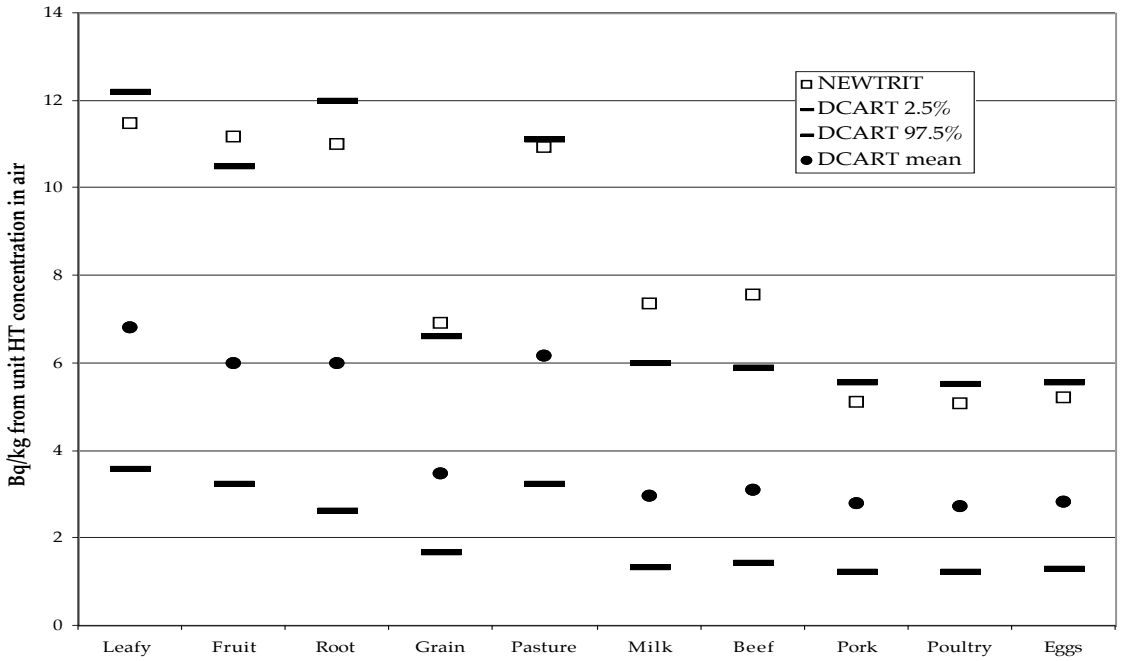

Figure 2. Concentrations of tritium in foodstuffs from a unit air concentration of HT predicted deterministically by NEWTRIT and stochastically by DCART.

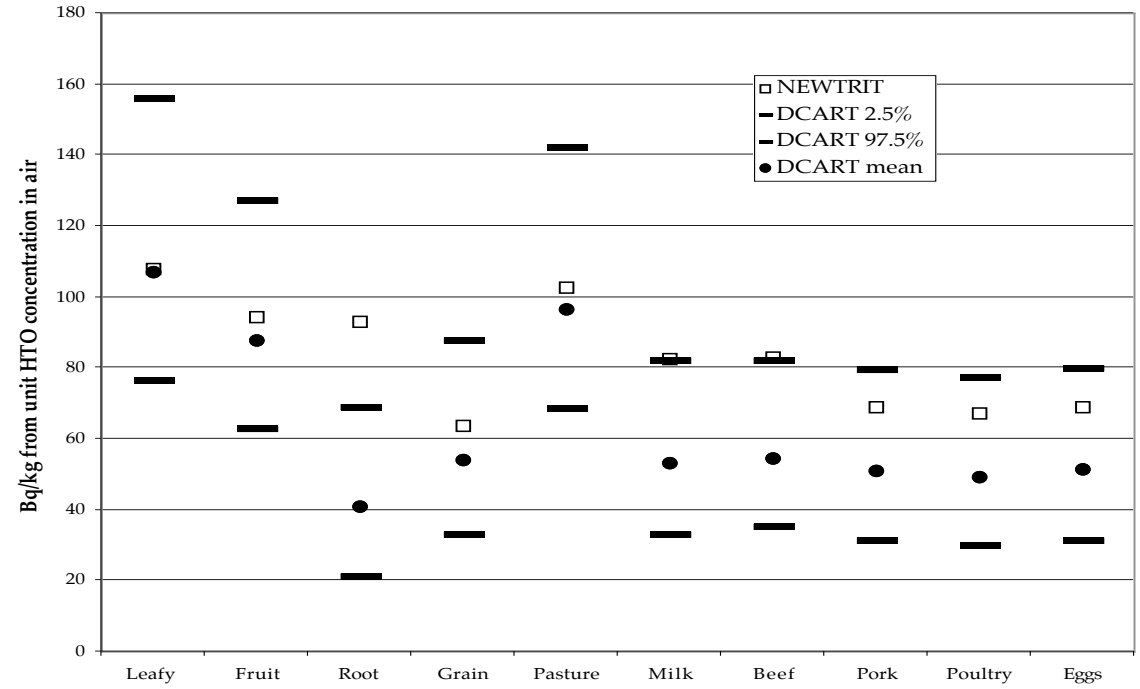

Figure 3. Concentrations of tritium in foodstuffs from a unit air concentration of HTO predicted deterministically by NEWTRIT and stochastically by DCART.

In Figure 2 (for unit air concentration of HT), the inherent conservatism of NEWTRIT stands out. NEWTRIT's prediction is either above the 97.5 percentile of the DCART prediction or is not far below it. Parameters for transfer of HT in the environment were chosen deliberately high so that NEWTRIT would be more acceptable to regulators requiring that a release of HT be modeled as if it were a release of HTO. Concentrations in vegetables are much lower in DCART than NEWTRIT because DCART is calibrated to an HTO in air moisture to HT in air volume ratio of 6.0, while NEWTRIT is calibrated to 8.0. Furthermore, the plant water concentrations in NEWTRIT are set at 1.5 times air moisture, while in DCART, plant water concentrations are only about 1.1 times air moisture. The large difference between DCART's mean concentration for root crops and NEWTRITs is due to the fact that DCART assumes that the root crop is nearly in equilibrium with the soil water, which is 30% the concentration of air moisture. In contrast, NEWTRIT, having no soil compartment, assumes conservatively that the concentration in root crops is the same as that of fruit vegetable.

In Figure 3 (for unit air concentration of HTO), the mean of DCART's stochastic predictions is not very different than NEWTRIT's prediction for vegetables, with the exception of root crops (explained above). These differences are accounted for by differences in assumptions about the magnitude of the transfer from air moisture to plant water (a fraction of 0.9 for NEWTRIT but variable and reduced in DCART because of dilution due to the soil water contribution) and differences in discrimination factors (0.9 or 0.8 in NEWTRIT depending upon the vegetable, and 0.7 in DCART).

NEWTRIT's higher predictions for animal products, obvious in both Figure 2 and Figure 3, are due to the conservative assumption in NEWTRIT that the concentration of tritium in the drinking water of animals is 50% that of air moisture (compared with 30% in DCART). NEWTRIT's concentrations of milk and beef, that lie above DCART's 97.5 percentile confidence limit, are due to a diet that is primarily pasture grass, rather than the mixture of pasture grass, hay and grain that is found in DCART.

CONCLUSION

DCART is a steady-state spreadsheet model for transfer of tritium from the atmosphere to dose. Developed at LLNL, DCART is adaptable, flexible, and transparent. It can be modified easily to account for new information. All doses are predicted as the mean and 95% confidence interval. DCART has been tested against observations and predictions of other models within international model validation groups. Many of the pathways are still untested because of the dearth of observational data; when transfers are not known well (as for animals), DCART errs on the conservative side.

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APPENDIX - All Parameters and Distributions in DCART

Table A1. Input parameters for the precipitation pathway; specific input required for specific sources and receptors. Generic values are provided as rough guidance. (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Source	Recommended value	Type of distribution	Generic values
Release rate	Bq/s	Facility records	Year-dependent	normal	NA
Distance source to receptor	m	GPS measurements	Receptor -specific	NA	NA
Fraction of year that it rains		15 minute data for 1997 - 2003	Annual, if known	normal	0.018 ± 0.00060
Washout coefficient	/s	Belot (1998)	For specific source to receptor	lognormal (GM ± GSD)	1.6 E-4 ± 2.0
Average rainfall rate when wind towards receptor from specific source[1]	mm/h	Mean of 1997 – 2003 data or year specific	For specific source to receptor	lognormal	2.1 ± 0.14
Wind speed while raining at approx. height of release	m/s	Mean of 1997 – 2003 data or year specific	For specific source to receptor	normal	2.8 ± 0.37 (10 m) 3.9 ± 0.41 (40 m)
Fraction of time wind into sector when raining		Mean of 1997 – 2003 data or year specific	For specific source to receptor	normal	See Table A8
Precipitation	m/y	LLNL annual records	Known annual	lognormal	0.34 ± 0.12 for 1952 - 2003
□ □	radians	LLNL wind rose	0.393		

[1] This parameter is not used directly in DCART; however, it is used to estimate the washout coefficient.

Table A2. Driving parameters: air concentrations

Parameter	Units	Source	Recommended value	Type of distribution	Range of values
HT or HTO concentration in air	Bq/m^3	Dispersion model (e.g., CAP88-PC)	Year-dependent;	normal	Year specific
HTO concentration in air	Bq/L or Bq/m^3	Annual average observed air tritium sampling locations	Year-dependent	normal	Year specific

Table A3. Input parameters for soil pathways (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Source	Recommended value	Type of distribution	Range of values
Dry deposition velocity, HTO	m/s	Tritium Data Base[a]	$5.3\ 10^{-3}$	lognormal (GM ± GSD)	$5.3\ 10^{-3} \pm 3.8$
Dry deposition velocity, HT	m/s	Tritium Data Base[a]	$2.\ 6\ 10^{-4}$	lognormal (GM ± GSD)	$2.6\ 10^{-4} \pm 3.5$
Fraction HTO retained by soil	Bq L^{-1} soil HTO / Bq L^{-1} air moisture	IAEA 2001	0.3 x air moisture	triangular	0.1 – 0.3 – 0.5
Fraction HTO in soil from HT release	Bq L^{-1} HTO / Bq m^{-3} HT in air	Davis and Bickel 2000	6.0 x HT in air	lognormal (GM ± GSD)	6.0 ± 1.5

[a] The Tritium Data Base contains experimental values of deposition velocities that have been published in the open literature.

Table A4. Input parameters for plant pathways (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Source	Recommended value	Type of distribution	Range of values
Absolute humidity	kg/m^3	Silica gel data; 1974 – 2003	Year-specific or 0.0079	normal	0.0078± 0.00040
Relative humidity		1999, 2001 - 2003 met data	Year-specific or 0.69	normal	0.69 ± 0.07
HTO concentration in plant water from HT	Bq/L HTO / Bq/m^3 HT	Davis and Bickel 2000	6.0	lognormal (GM ± GSD)	6.0 ± 1.5
HTO/H_2O ratio of vapor pressure	g	Raney and Vaadia 1965	0.909		
Dry mass of: Leafy vegetables Fruit, Fruit vegs Root crops Grain Pasture –fresh Hay Tree Rings	kg dry weight / kg fresh weight	Ciba-Geigy 1988	 0.094 0.136 0.15 0.882 0.253 0.902 1.0	 uniform extreme value dist lognormal (GM ± GSD) normal uniform extreme value dist.	 0.03 – 0.15 m = 0.11; scl = 0.05 0.14 ± 1.56 0.88 ± 0.02 0.18 – 0.32 md = 0.89; scl = 0.01
Water equivalent factor: Leafy vegetables Fruit, Fruit vegs. Root crops Grain Pasture Hay Tree rings	L / kg	Calculated from fresh weights, dry weights, and hydrogen content; Ciba-Geigy 1981 Measured, CRL	 0.6 0.581 0.575 0.577 0.590 0.583 0.57	 lognormal (GM ± GSD) lognormal (GM ± GSD) logistic uniform uniform uniform	 0.600 ± 1.03 0.581 ± 1.02 m = 0.575; s = 0.003 0.566 – 0.581 0.586 – 0.594 0.580 – 0.587
Isotopic discrimination		Kim & Baumgärnter 1994; Kalin et al 1995	0.7 (plants) 0.4 (tree rings)	extreme value dist. triangular	md= 0.67; scl = 0.14 0.2 - 0.4 - 0.7
Fraction of tritium from air moisture			0.6 (fruit and grain) 0.05 (root crops)	Triangular triangular	0.5 – 0.6 –0.7 0.0 –0.05 –0.20

Table A5. Input parameters for animal pathways. (Correlations: -0.9 pasture with hay; -0.9 pasture with grain; 0.90 and 0.95 grain intake of pigs and poultry, respectively, with water intake) (GM = geometric mean; GSD = geometric standard deviation)

Parameter	Units	Source	Recommended value	Type of distribution	Range of values
Diet, pasture: Milk cow Beef cow	kg fresh weight / d	NAS 1996; 2001;	 25.0 13.0	 normal normal	 25 ± 5.0 13 ± 2.6
Diet, hay: Milk cow Beef cow	kg fresh weight / d	NAS 1996; 2001;	 11.6 6.2	 normal normal	 11.6± 2.32 6.2 ± 0.88
Diet, grain: Milk cow Beef cow Pigs Chicken Laying hen	kg fresh weight / d	NAS 1994; 1996; 1998; 2001;	 3.0 1.5 3.4 0.18 0.11	 normal normal normal normal normal	 3.0 ± 1.0 1.5 ± 0.5 3. 34 ± 0.34 0.18 ± 0.021 0.11 ± 0.017
Concentration of animal drinking water: fraction of air		Empirical data from LLNL pool, 1990 - 2000	0.165	lognormal (GM ± GSD)	0.16 ± 1.5
Water intake: Milk cow Beef cow Pig Chicken Laying hen	L / d	NAS 1994; 1996; 1998; 2001;	 103. 44. 8.5 0.36 0.22	 normal normal normal normal normal	 103 ± 1.82 44.3 ± 1.9 7.5 ± 1.1 0.35 ± 0.042 0.22 ± 0.034

Table A5 continued

Parameter	Units	Source	Recommended value	Type of distribution	Range of values
Inhalation rate: Milk cow Beef cow Pigs Chicken Laying hen	m^3/d	Assorted values, mostly from models	 158 127 43.0 1.0 1.0	 truncated normal truncated normal truncated normal truncated normal truncated normal	 144 ± 67.0 (75 – 300) 127 ± 72.0 (60 – 300) 43 ± 18 ((20 – 70) 1.0 ± 0.60 (0.3 – 2.) 1.0 ± 0.60 (0.3 – 2.)
Water from inhalation	kg/d	Raskob 1994	1.5 x inhalation rate (m^3/d) x absolute humidity (kg/m^3)		
Dry mass of: Milk Beef Pork Chicken Egg	kg dry weight / kg fresh weight	Ciba Geigy 1981	 0.103 0.332 0.5 0.33 0.26	 uniform triangular custom distrib uniform uniform	 0.09 – 0.12 0.28 - 0.32 - 0.44 70% 0.28-0.48; 30% 0.8 – 1 0.27 – 0.39 0.25 – 0.27
Water equivalent: Milk Beef Pork Chicken Eggs	L/kg	Calculated from fresh weights, dry weights, and hydrogen content; Ciba-Geigy 1981	 0.711 0.795 0.904 0.796 0.835	 triangular triangular uniform uniform uniform	 0.592 – 0.711 – 0.746 0.72 – 0.8 – 0.90 0.786 – 1.0 0.73 – 0.85 0.834 – 0.836
Uncertainty on transfer to animal OBT			1.0	truncated normal	1.0 ± 0.4; (0.8 - ∞)

Table A6. Inhalation (Myers et al. 2000) and ingestion intake rates (EPA 1999) for adult, child (age 10) and infant (6m to 1y); correlations: -0.75 leafy vegetables and fruit/fruit vegetables; -0.8 root crops and grain; -0.8 beef and pork. (GM= geometric mean, GSD = geometric standard deviation)

Parameter	Units		Recommended value	Type of distribution	Range of values
Inhalation rate	m^3/y	Adult Child Infant	4860. 4930. 1640.	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	4604 ± 1.37 4689 ± 1.37 1346 ± 1.87
HTO concentration in air from HT	Bq/L HTO / Bq/m^3 HT	Davis and Bickel 2000	4.0 for inhalation	lognormal (GM ± GSD)	4.0 ± 1.5
Drinking water	L/a	Adult Child Infant	552. 356. 120.	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	510 ± 1.55 318 ± 1.66 107± 2.03
Leafy vegetables	kg/y	Adult Child Infant	15.9 9.53 1.17	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	11.0 ± 2.37 6.51 ± 2.40 0.43 ± 4.12
Fruit, fruit vegetables	kg/y	Adult Child Infant	117. 107. 64.8	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	99.8 ± 2.2 92.4 ± 1.79 46.4 ± 2.23
Root crops	kg/y	Adult Child Infant	27.6 24.5 6.17	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	20.5 ± 2.32 16.3 ± 2.62 0.61 ± 10.1
Grain	kg/y	Adult Child Infant	80.0 85.1 23.4	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	69.3 ± 1.78 75.5 ± 1.64 20.1 ± 2.04
Milk (and products)	kg/y	Adult Child Infant	91.2 177. 208.	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	69.1 ± 2.22 157 ± 1.71 152 ± 2.17
Beef	kg/y	Adult Child Infant	17.7 14.5 3.13	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	14.1 ± 2.12 12.2 ± 2.02 1.62 ± 3.06

Table A6 continued

Parameter	Units		Recommended value	Type of distribution	Range of values
Pork	kg/y	Adult Child Infant	5.68 4.66 0.967	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	1.83 ± 4.79 1.54 ± 4.58 0.259 ± 6.29
Chicken	kg/y	Adult Child Infant	12.9 11.5 2.71	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	7.98 ± 2.74 7.37 ± 2.70 0.591 ± 5.92
Eggs	kg/y	Adult Child Infant	6.24 5.49 2.63	lognormal (GM ± GSD) lognormal (GM ± GSD) lognormal (GM ± GSD)	4.90 ± 2.88 4.52 ± 3.12 2.89 ± 2.33

Table A7. Dose coefficients (ICRP (1995, 1996); uncertainty from Harrison et al. 2002) (GM= geometric mean, GSD = geometric standard deviation)

Parameter	Units		Recommended value	Type of distribution	Range of values
HT inhalation	Sv/Bq	Adult Child Infant	$1.8\ 10^{-15}$ $2.3\ 10^{-15}$ $4.8\ 10^{-15}$	lognormal (GM ± GSD)	$3.82\ 10^{-15}$ ± 1.23 $4.43\ 10^{-15}$ ± 1.32 $9.22\ 10^{-15}$ ± 1.33
HTO inhalation/ingestion	Sv/Bq	Adult Child Infant	$1.8\ 10^{-11}$ $2.3\ 10^{-11}$ $4.8\ 10^{-11}$	lognormal (GM ± GSD)	$3.82\ 10^{-11}$ ± 1.23 $4.43\ 10^{-11}$ ± 1.32 $9.22\ 10^{-11}$ ± 1.33
OBT ingestion	Sv/Bq	Adult Child Infant	$4.2\ 10^{-11}$ $5.7\ 10^{-11}$ $1.2\ 10^{-10}$	lognormal (GM ± GSD)	$8.34\ 10^{-11}$ ± 1.34 $1.02\ 10^{-10}$ ± 1.47 $2.22\ 10^{-10}$ ± 1.49

Table A8. Fraction of time the wind blows towards all sectors when raining (average of data 1997 – 2003). Uncertainty shown for directions from sources towards the Visitors Center.

Direction towards	10 m winds	40 m winds
N	0.079	0.096
NNE	0.14 ± 0.027	0.15 ± 0.024
NE	0.17	0.16
ENE	0.11 ± 0.031	0.12 ± 0.039
E	0.074	0.068
ESE	0.036 ± 0.012	0.030 ± 0.012
SE	0.019	0.015
SSE	0.019	0.020
S	0.036	0.033
SSW	0.036	0.040
SW	0.061	0.048
WSW	0.066	0.047
W	0.035	0.057
WNW	0.030	0.037
NW	0.026	0.028
NNW	0.059	0.055

Table A9. Parameters used in the sub-model for dose from swimming. (GM= geometric mean, GSD = geometric standard deviation)

Parameter	Units	Source		Recommended value	Type of distribution	Range of values
Ratio air moisture VIS/POOL air tritium samplers		Empirical from LLNL data		2.1535	normal	2.1535 ± 0.215
Ratio pool water/air moisture at POOL		Empirical from LLNL data		0.165	lognormal (GM ± GSD)	0.16 ± 1.5
Skin area	m^2	EPA 1999	Adult Child Infant	1.82 1.12 0.425	normal	1.82 ± 0.25 1.12 ± 0.12 0.425 ± 0.045
Humidity at skin/water temperature	mg/L	Weight of saturated water vapor		0.2724	triangular	0.0224 – 0.02724 – 0.2878
Intake rate for skin	L/min m^2	Osborne 1968		5.1	normal	5.1 ± 1.5
Minutes spent swimming per year		EPA 1999	Adult Child Infant	150 720 510	triangular	0 – 150 – 9000 0 – 720 – 2500 0 – 510 - 2500